Emissions Control for Lean Gasoline Engines

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This presentation does not contain any proprietary, confidential, or otherwise restricted information

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  - Chris Owens, Ken Price, Tom Pauly, Doug Ball, Davion Clark, David Moser, Corey Negohosian

Project Overview

Timeline

- Year 2 of 3-year program
  - **Project start date:** FY2016
  - **Project end date:** FY2018
- Builds on previous R&D in FY13-FY15

Budget

- FY16: $400k (Task 2*)
- FY17: $400k (Task 2*)

*Task 2: Lean Gasoline Emissions Control

Part of large ORNL project "Enabling Fuel Efficient Engines by Controlling Emissions" (2015 VTO AOP Lab Call)

Barriers Addressed

- Barriers listed in VT Program Multi-Year Program Plan:
  - 2.3.1B: *Lack of cost-effective emission control*
  - 2.3.1C: *Lack of modeling capability for combustion and emission control*
  - 2.3.1.D: *Emissions control durability*

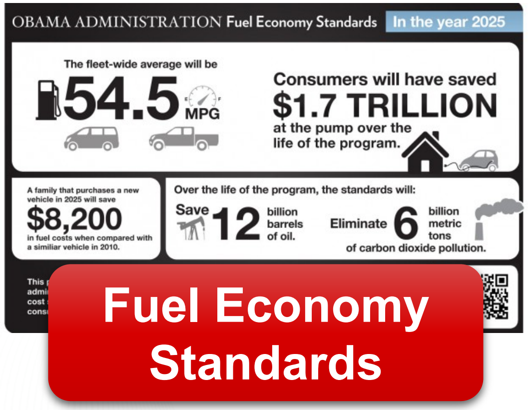
Collaborators & Partners

- General Motors
- Umicore
- University of South Carolina
- Cross-Cut Lean Exhaust Emissions Reduction Simulations (CLEERS)

Objectives and Relevance

Enabling lean-gasoline vehicles to meet emissions regulations will achieve significant reduction in petroleum use

- Objective:
  - Demonstrate technical path to emission compliance that would allow the implementation of lean gasoline vehicles in the U.S. market.
    - Lean vehicles offer 5–15% increased efficiency over stoichiometric-operated gasoline vehicles



54.5 mpg CAFE by 2025

Objectives and Relevance

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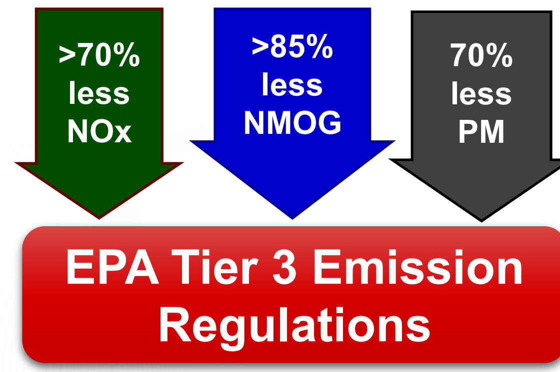
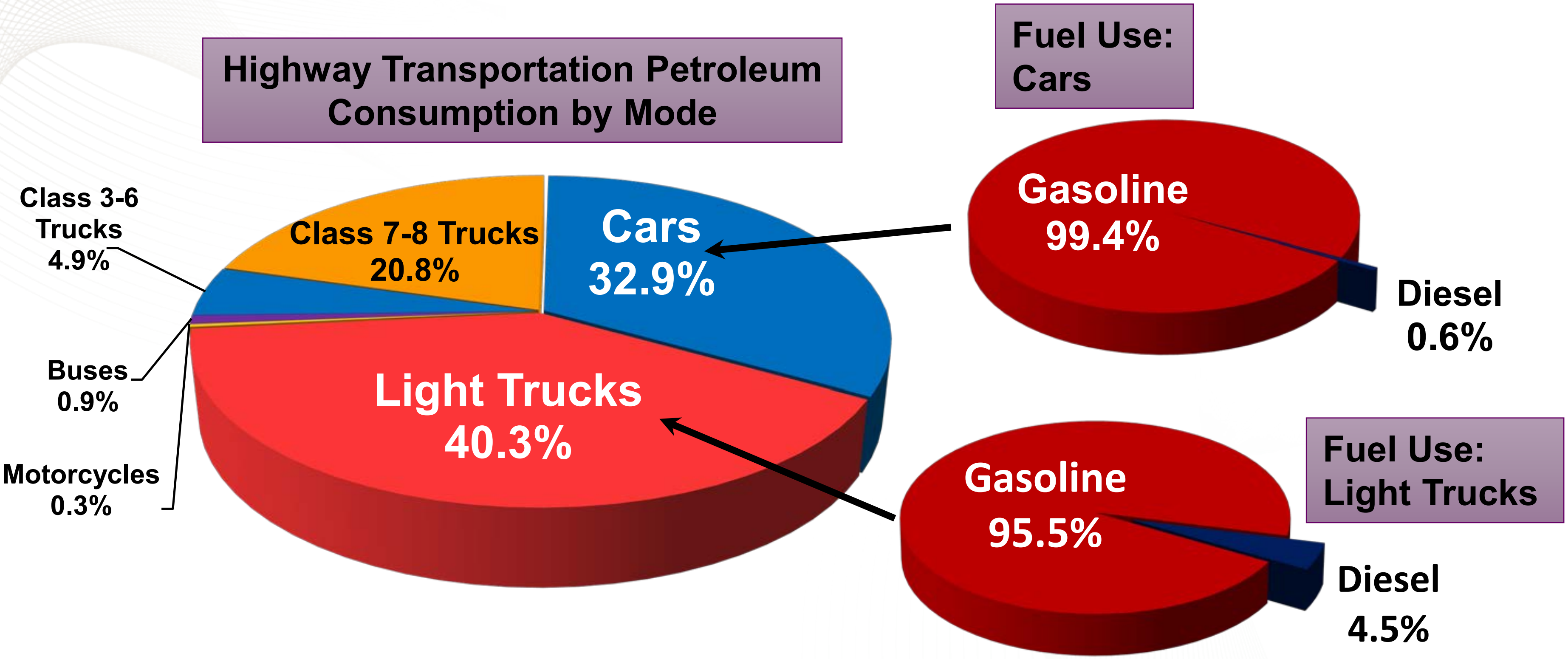
- <u>Objective:</u>
  - Demonstrate technical path to emission compliance that would allow the implementation of lean gasoline vehicles in the U.S. market.
    - Lean vehicles offer 5–15% increased efficiency over stoichiometric-operated gasoline vehicles
    - Compliance required: U.S. EPA Tier 3
  - Investigate strategies for cost-effective compliance
    - minimize precious metal content while maximizing fuel economy



- <u>Relevance:</u>
  - U.S. passenger car fleet is dominated by gasoline-fueled vehicles.
  - Enabling introduction of more efficient lean gasoline engines can provide significant reductions in overall petroleum use
    - thereby lowering dependence on foreign oil and reducing greenhouse gases

Relevance: small improvements in gasoline fuel economy <u>significantly</u> decreases fuel consumption



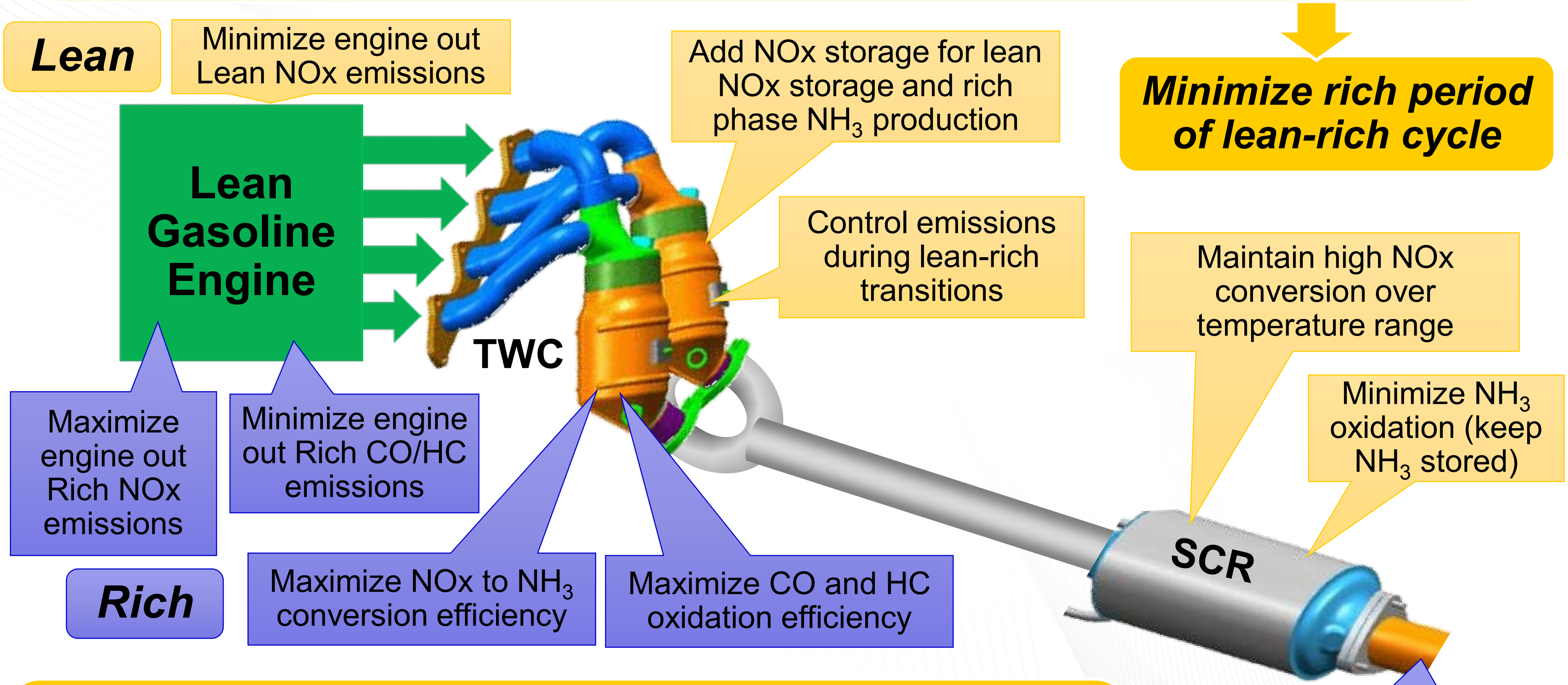
- US car and light-truck fleet dominated by gasoline engines
- 10% fuel economy benefit has significant impact
  - Potential to save 13 billion gallons gasoline annually
- HOWEVER…emissions compliance needed!!!

Lean gasoline vehicles can decrease US gasoline consumption by <u>~13 billion gal/year</u>

Reference: 2016 Transportation Energy Data Book, Ed. 35 (2014 data)

Approach focuses on catalyst and system optimization of Passive SCR (and LNT+SCR)

Key Principle: system fuel efficiency gain depends on optimizing NH_3 production during rich operation and NOx reduction during lean operation

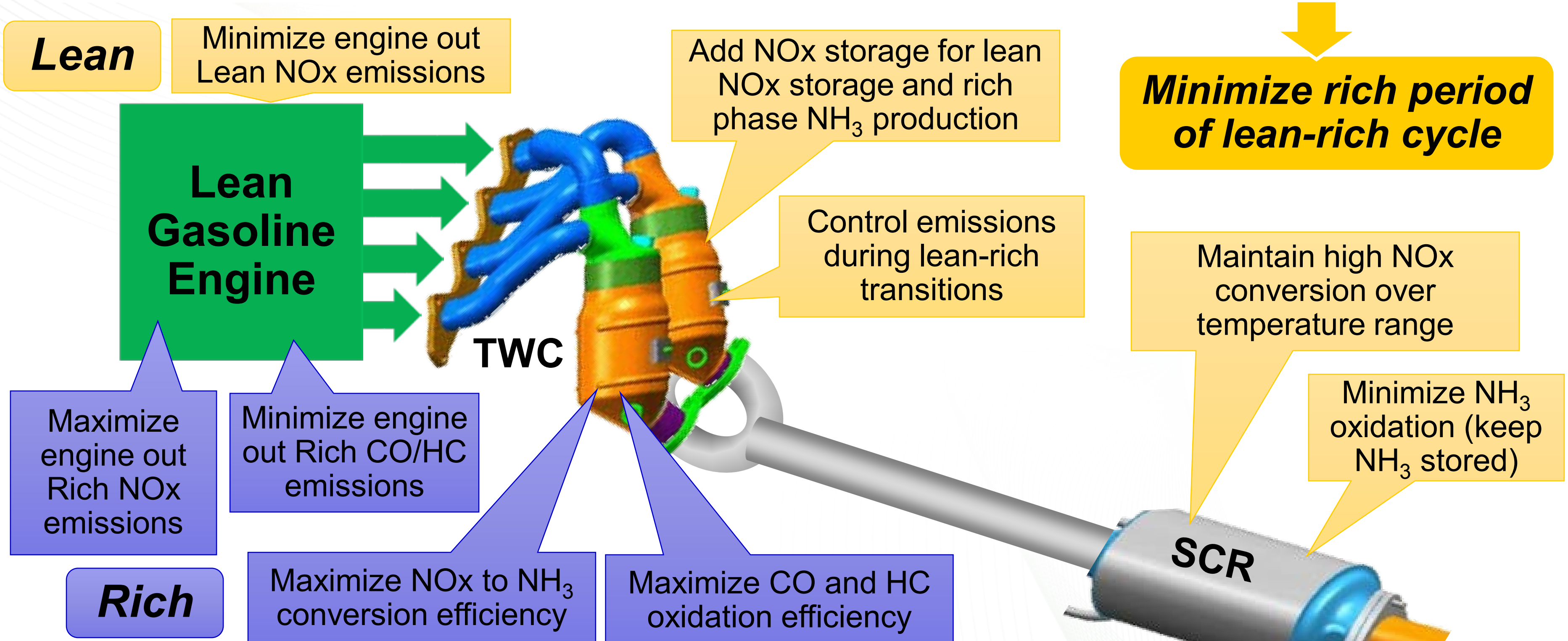


Other Core Principles:

- Expand range of temperature operation
- Materials must be durable to temperature and poisons (S)
- Understand Pt group metals utilization to minimize cost

Approach focuses on catalyst and system optimization of Passive SCR (and LNT+SCR)

Key Principle: system fuel efficiency gain depends on optimizing NH_3 production during rich operation and NOx reduction during lean operation



References to NH_3 production/utilization in Passive SCR, LNT+SCR, HC-SCR, etc.:
W. Li, K. L. Perry, K. Narayanaswamy, C. H. Kim and P. Najt, SAE 2010-01-0366 (GM)
Y. Liu, M.P. Harold, D. Luss, *Appl. Cat. B*, **121–122**, 239–251.
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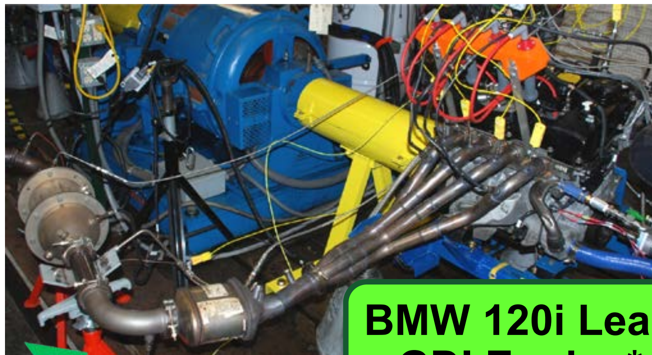
J. Theis, M. Dearth, R. McCabe, SAE 2011-01-0305 (Ford)
V. Easterling, Y. Ji, M. Crocker, M. Dearth, R.W. McCabe, *Appl. Cat. B* **123–124**, 339–350.
J. A. Pihl, J. E. Parks, C.S. Daw, and T.W. Root, SAE 2006-01-3441
J. Parks and V. Prikhodko, SAE 2009-01-2739
C. L. DiMaggio, G. B. Fisher, K. M. Rahmoeller, and M. Sellnau, SAE 2009-01-0277

Approach Combines Engine, Bench, and Aging Studies to Achieve Project Goals

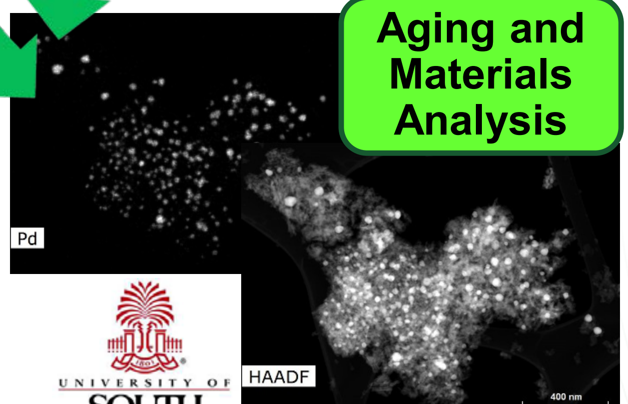
GM *System Study Guidance*



BMW 120i Lean GDI Engine*

Bench Flow Reactor

umicore Automotive Catalysts *Prototype Catalysts*



Aging and Materials Analysis

UNIVERSITY OF SOUTH CAROLINA *Hydrothermal and S Aging*

Micrographs were obtained using instrumentation (FEI Talos F200X S/TEM) provided by the Department of Energy, Office of Nuclear Energy, Fuel Cycle R&D Program and the Nuclear Science User Facilities.

Project Goals

Fuel Efficiency Gain
(vs. Stoichiometric GDI case)
and Greenhouse Gas Emissions

Tier 3 Emissions
NOx, CO, HCs, PM

Tier 3 Durability (150k miles)
Hydrothermal and S Aging

Cost-Effectiveness
Minimize PGM and complexity

*Lean GDI engine has full pass controller (National Instruments Powertrain)

Collaborations and Partners

Primary Project Partners

- **GM**
  - guidance and advice on lean gasoline systems via monthly teleconferences
- **Umicore**
  - guidance (via monthly teleconferences) and catalysts for studies (both commercial and prototype formulations)
- **University of South Carolina (Jochen Lauterbach)**
  - Catalyst aging studies with student Calvin Thomas

Additional Collaborators/Partners on Project/Engine Platform (Since Project Inception)

- **CDTi:** catalysts for studies
- **CLEERS:** Share results/data and identify research needs
- **LANL:** Engine platform used for NH_3 sensor study (Mukundan, Brosha, Kreller)
- **MECA:** GPF studies via Work For Others contract
- **University of Minnesota:** Collaboration on DOE funded project at U of Minn. related to lean GDI PM (PI: Will Northrop)
- **CTS (Filter Sensing Technologies):** Small business technical assistance on RF sensors for GPF on-board diagnostics
- **Tennessee Tech University:** Project data being used for lean gasoline emission control system modeling
- **DOE VTO Fuel and Lubricant Technology Program:** Engine platform used for ethanol-based HC-SCR studies

R&D Expanded Coverage via Collaborations:

- Lean GDI PM Control
- Sensors
- Modeling
- Fuels

Responses to 2016 Reviewers

FY2016 AMR Review
(3 Reviewers)
[scores: 1 (min) to 4 (max)]

Category	Score
Approach	3.33
Tech Accomplishments	3.67
Collaboration	3.83
Future Research	3.50
Weighted Average	**3.58**

Relevant to DOE Objectives?
YES (100%)

Sufficiency of Resources	
Insufficient (33%)	Sufficient (67%)

Summary of Reviewers' Feedback:

- Generally positive feedback on:
  - Approach (bench+engine+aging)
  - Collaborations with industry
- Interest in engines with lower NOx/temperature
- Interest in more utilization of engine controls
- Interest in multi-step rich event and transients
- HC control is a priority (real challenge)
- Interest in H_2 influence
- Interest in SCR on filter and low N_2O SCR*

Project Adjustments/Responses:

- Many results translate to newer engines
- Variable valve and control research expanded
- Rich tip-in effects studied (ongoing)
- Agree: HC a real challenge (continued focus)
- Aging studies understanding H_2 influence
- Next priority for SCR is aging

*N_2O Greenhouse Gas (GHG) cap is 10 mg/mile

Milestones

Quarterly Milestones

Complete
- **FY2016, Q1:** Complete bench flow reactor assessment of Pd-only and TWC/NSC formulations for NH_3 production during Passive SCR

Complete
- **FY2017, Q3:** Evaluate three commercial or commercial-intent SCR catalyst formulations under dynamic air/fuel ratio operation relevant to lean gasoline engine application.

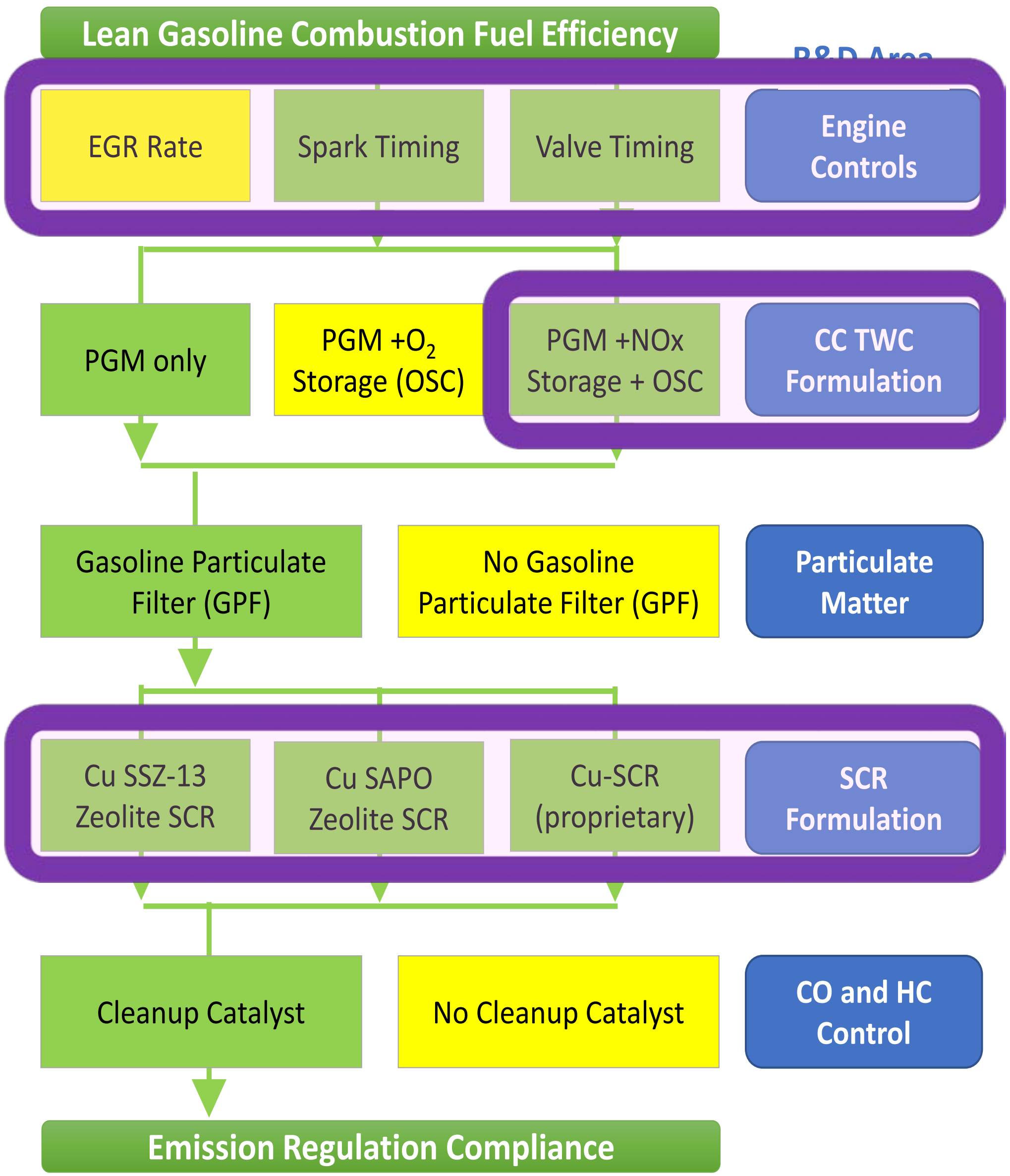
Annual SMART Milestones

On Track
- **FY2017:** (SMART) Meet EPA Tier 3 emission levels with a lean GDI engine while using less than 4 g Platinum Group Metal per liter of engine displacement (cost-related metric) and determine fuel efficiency benefit over USDRIVE naturally aspirated gasoline engine baseline efficiency at eight speed and load points defined by industry collaborators GM and Umicore. Based on drive cycle modes, determine which speed and load points are feasible for lean operation.

GO/NO-GO Decision

Complete
- **FY2017, Q2:** Demonstrate a pathway to an emissions control system that enables a lean gasoline direct injection engine to achieve U.S. EPA Tier 3 emission levels thereby enabling commercial viability of this petroleum saving lean gasoline engine technology.

Go/No-Go Decision Point: Pathway Defined



Key

- Feasible
- Limited Benefits
- Proven Not Possible

Project Goals

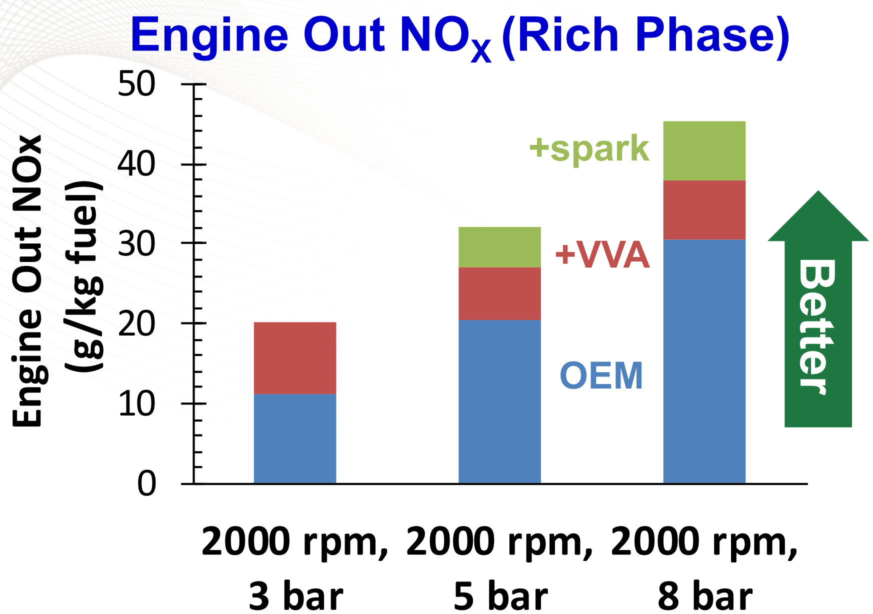
Fuel Efficiency Gain
(vs. Stoichiometric GDI case)
and Greenhouse Gas Emissions

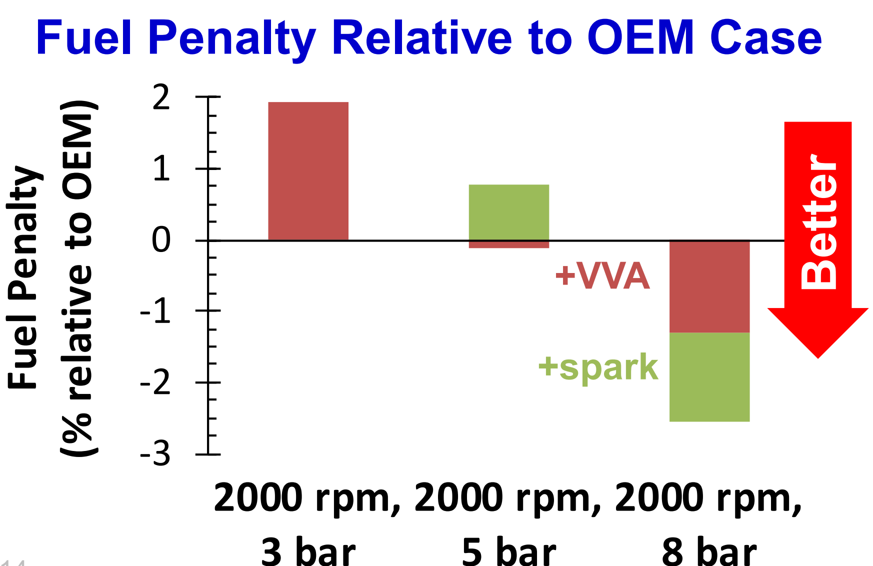
Tier 3 Emissions
NOx, CO, HCs, PM

Tier 3 Durability (150k miles)
Hydrothermal and S Aging

Cost-Effectiveness
Minimize PGM and complexity

Engine controls can increase NH_3 production: net benefits vary with engine load





Goal: maximize engine out Rich NOx emissions for NH_3 production

- Three engine combustion control strategies have been studied at 3, 5, and 8 bar for optimizing passive SCR
  - EGR rate
  - Spark Timing
  - Variable Valve Actuation (VVA)
- Spark Timing and VVA enable higher engine out NOx rates during the rich phase of the lean-rich cycle
  - more NOx=more NH_3=less fuel
  - Spark timing and VVA benefits can be combined for additive benefit
  - At 8 bar, further benefits occur with greater combustion efficiency

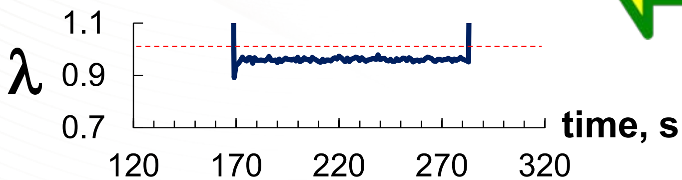
Spark Timing + VVA gives greater rich combustion *and* system fuel efficiency

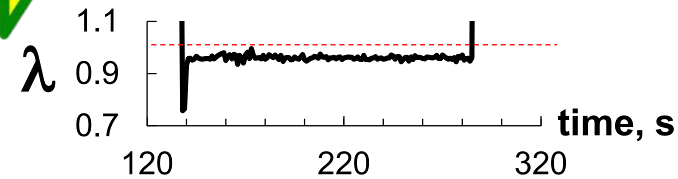
Sharp increase in NH_3 production early in rich period due to NOx storage component <u>and</u> rich "tip-in"

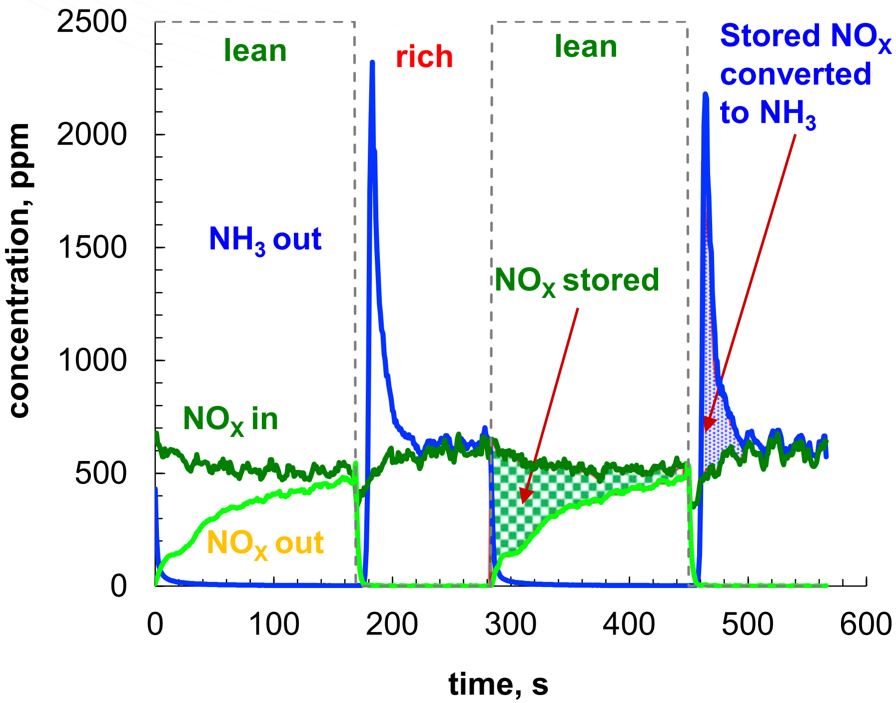
TWC with O_2 and NOx Storage Components
ORNL-1*

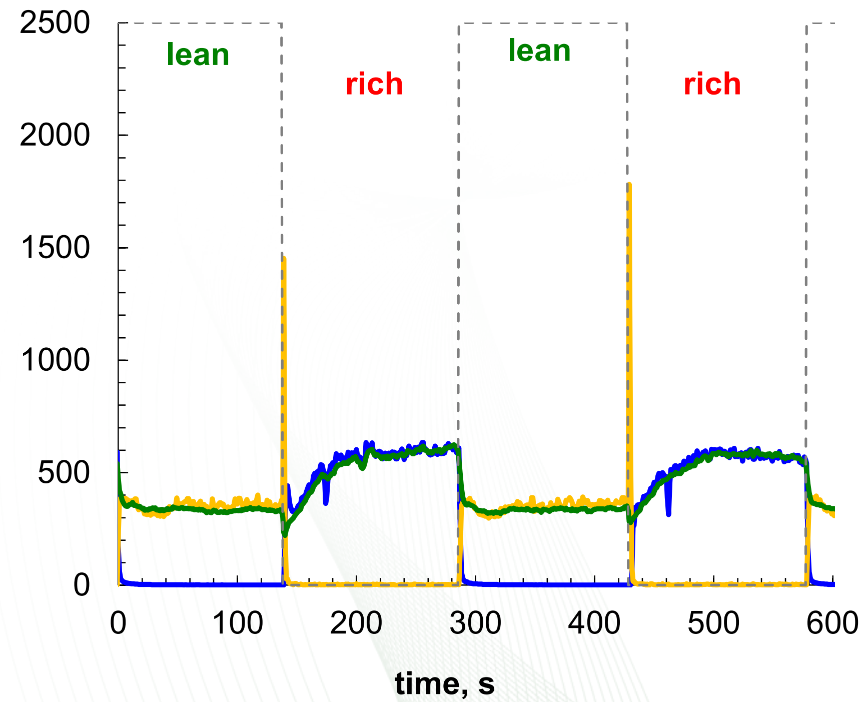
Catalyst Comparison

TWC with Pd only (no O_2/NOx storage)
Malibu-1*









*lean: 2000 rpm 3 bar; λ 1.77; **45000 h^{-1}**; TWC avg Tin=395 ºC*
*rich: 2000 rpm 3 bar; λ 0.96; **30000 h^{-1}**; TWC avg Tin=413 ºC*

*lean: 2000 rpm 2 bar; λ 2.0; **42000 h^{-1}**; TWC avg Tin=459 ºC*
*rich: 2000 rpm 2 bar; λ 0.96; **28000 h^{-1}**; TWC avg Tin=463 ºC*

*complete details of TWC formulations in technical back-up slides

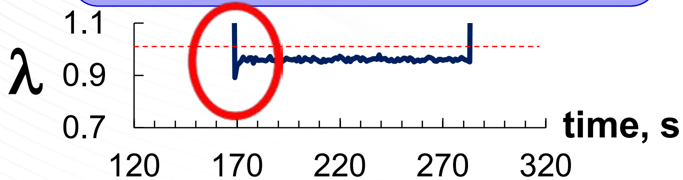
Sharp increase in NH_3 production early in rich period due to NOx storage component <u>and</u> rich "tip-in"

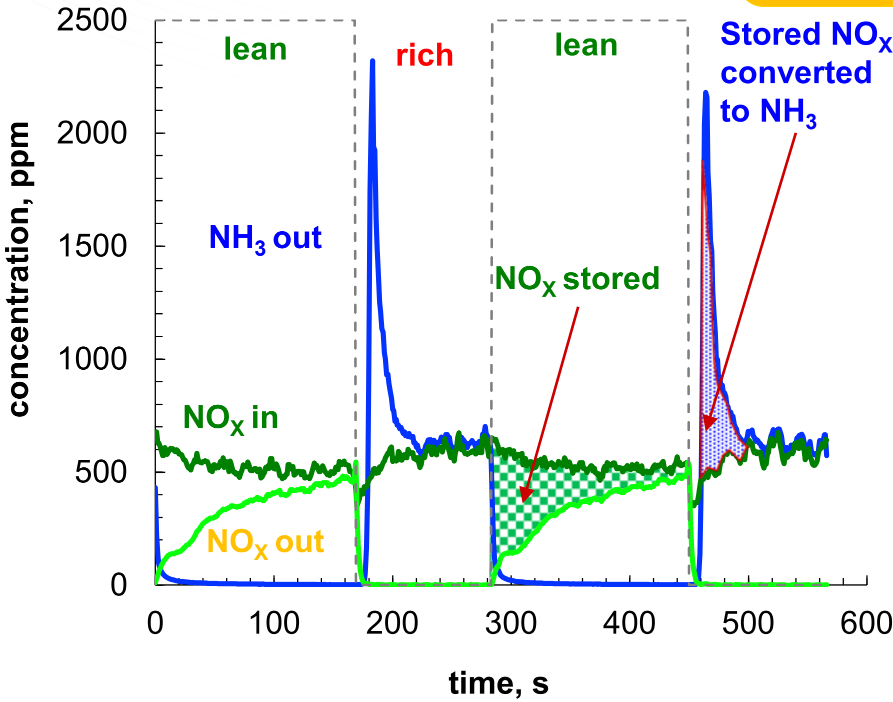
Engine Study
tip-in, rich overshoot

Engine vs. Bench Comparison

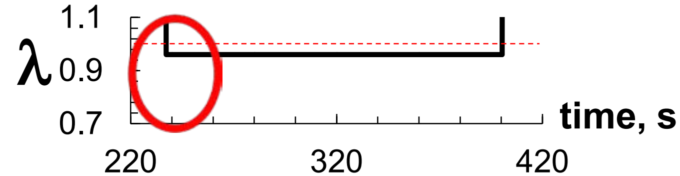
Bench Flow Reactor
no tip-in, square well λ trace

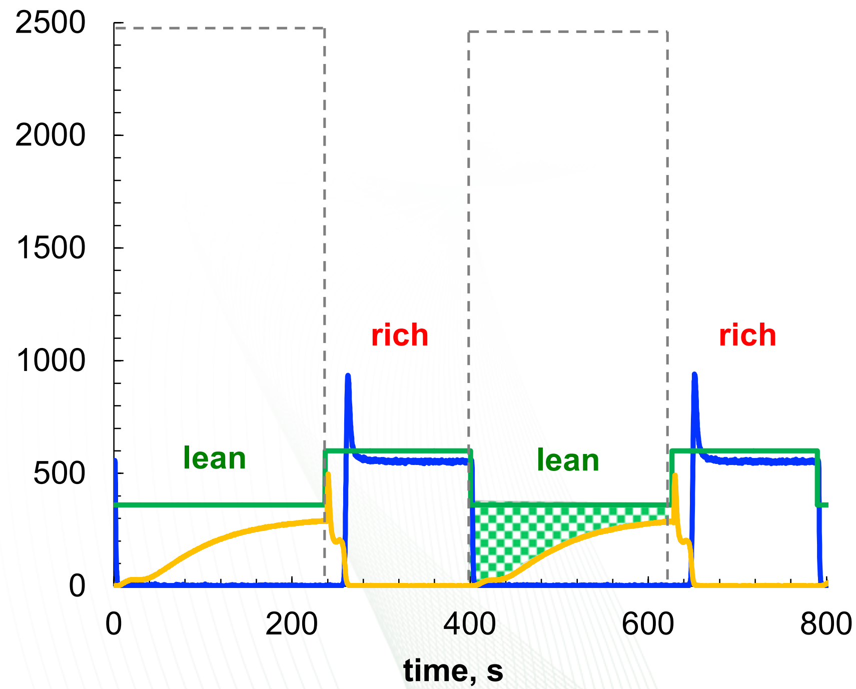
TWC with O_2 + NOx Storage
ORNL-1*





*lean: 2000 rpm 3 bar; λ 1.77; **45000 h^{-1}**; TWC avg Tin=395 ºC*
*rich: 2000 rpm 3 bar; λ 0.96; **30000 h^{-1}**; TWC avg Tin=413 ºC*





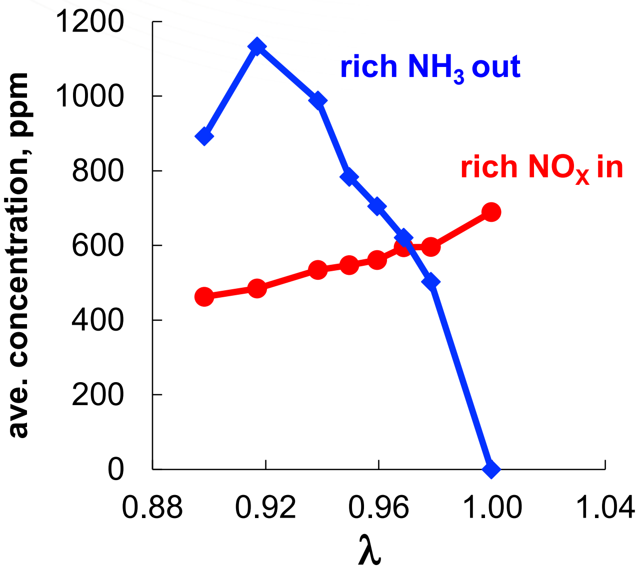
*lean: 2000 rpm 2 bar; λ 2.0; **45000 h^{-1}**; TWC Tin=400 ºC*
*rich: 2000 rpm 2 bar; λ 0.96; **27000 h^{-1}**; TWC Tin=400 ºC*

Also, see Theis et al. SAE 2015-01-1004, SAE 2015-01-1006

NH_3 production spike at rich onset only occurs when NOx is stored on the TWC [at high temperatures NOx not stored]

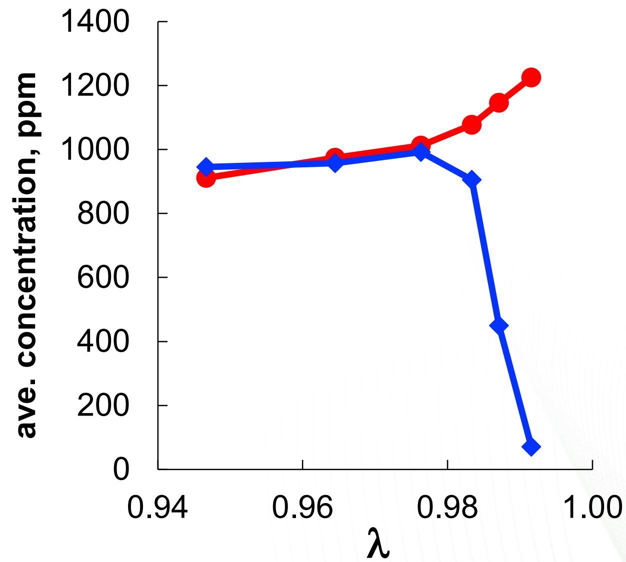
TWC with O_2 and NOx Storage Components
ORNL-1*

3 bar at 2000rpm
Average TWC_{inlet}=395°C



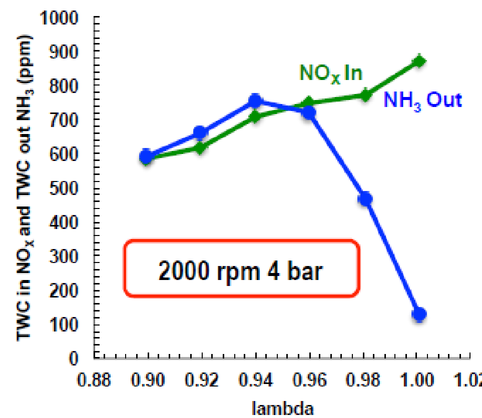
*lean: 2000 rpm 3 bar; λ 1.77; **45000 h^{-1}***
*rich: 2000 rpm 3 bar; λ 0.97; **30000 h^{-1}***

5 bar at 2000rpm
Average TWC_{inlet}=492°C



*lean: 2000 rpm 5 bar; λ 2.0; **55000 h^{-1}***
*rich: 2000 rpm 5 bar; λ 0.97; **40000 h^{-1}***

TWC with Pd only
(no O_2/NOx storage)
Malibu-1*



Previous data of Pd only TWC for comparison

*complete details of TWC formulations in technical back-up slides

Stored NOx on TWC helps enable increased NH_3 production for short (~2 sec) rich period operation

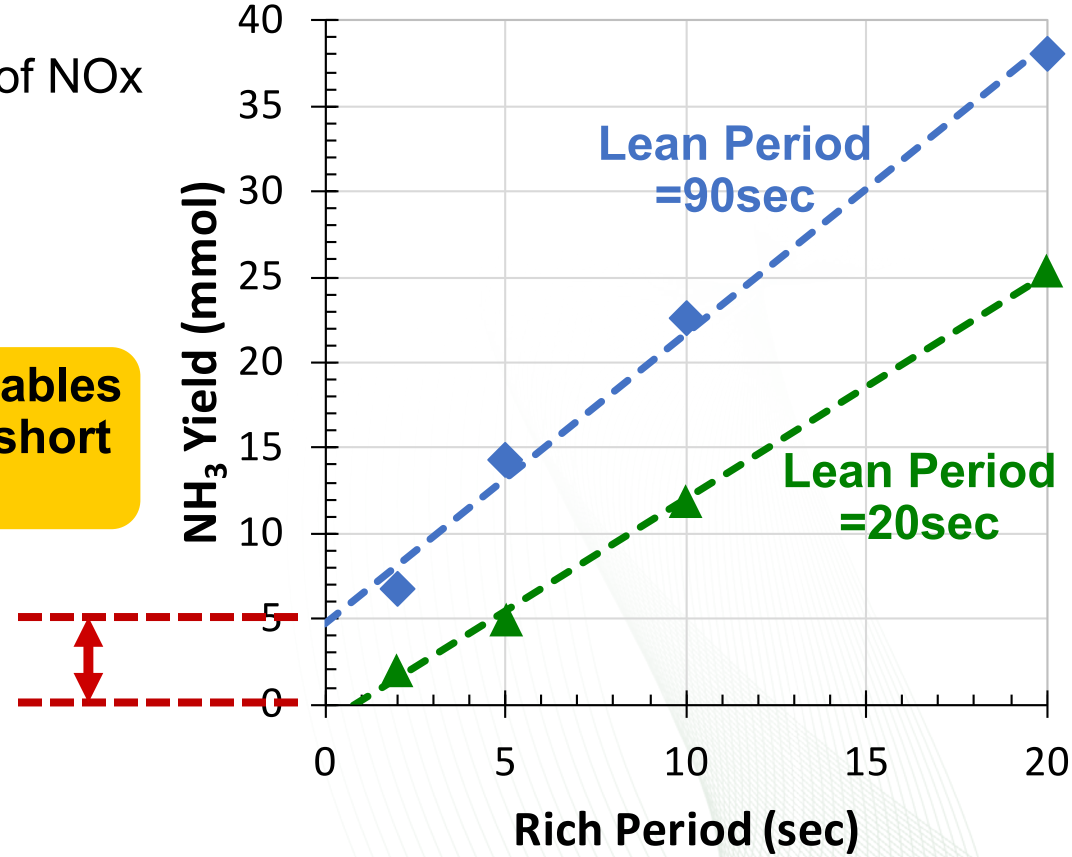
- Experiment investigated potential for NH_3 production under short rich periods representative of transient accelerations
- Compare different levels of NOx storage:
  - Lean Period = 90 sec
  - Lean Period = 20 sec

TWC with NOx storage enables better NH_3 production in short rich transients

Y-offset shows benefit of stored NOx for short rich period NH_3 production

TWC with O_2 and NOx Storage Components
ORNL-1*

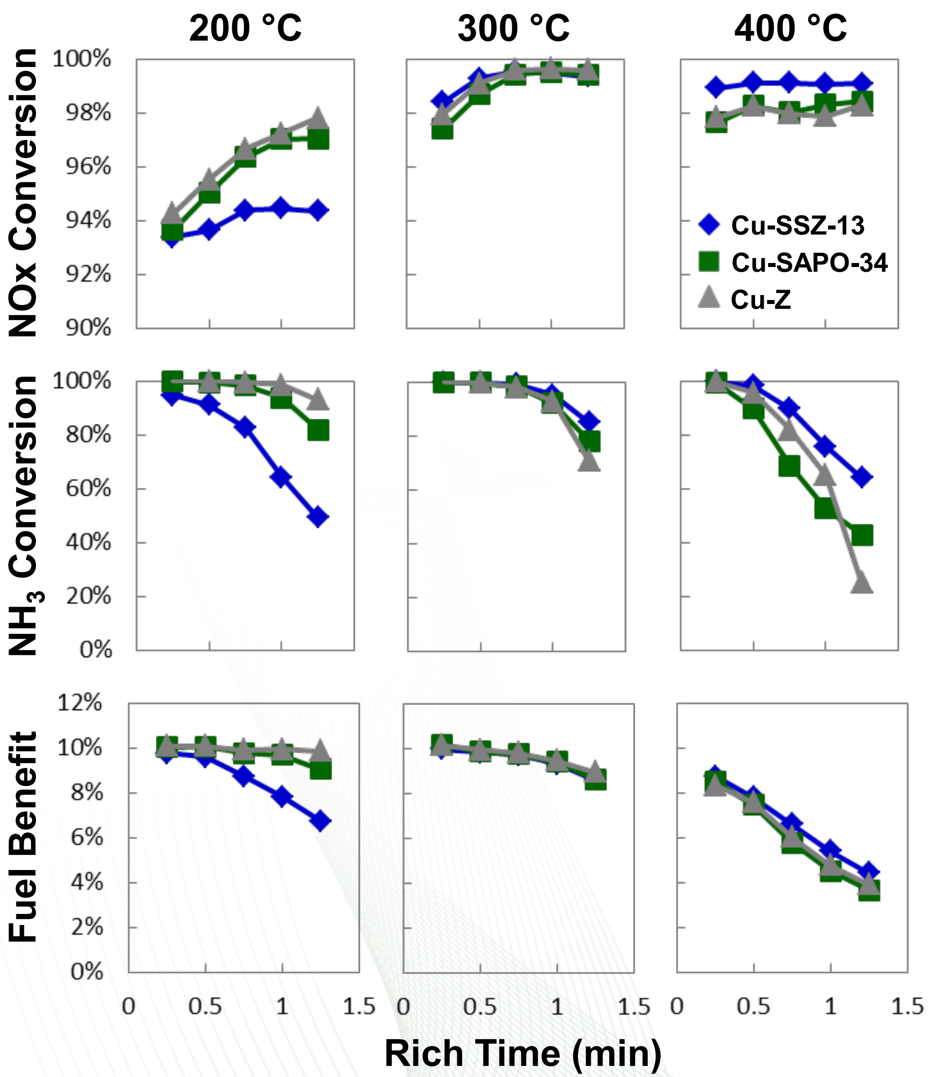
2000rpm, Lean 3 bar-Rich 8 bar, Rich λ=0.96



*complete details of TWC formulations in technical back-up slides

Small pore Cu zeolite formulation does not have a strong impact on passive SCR performance, but timing matters

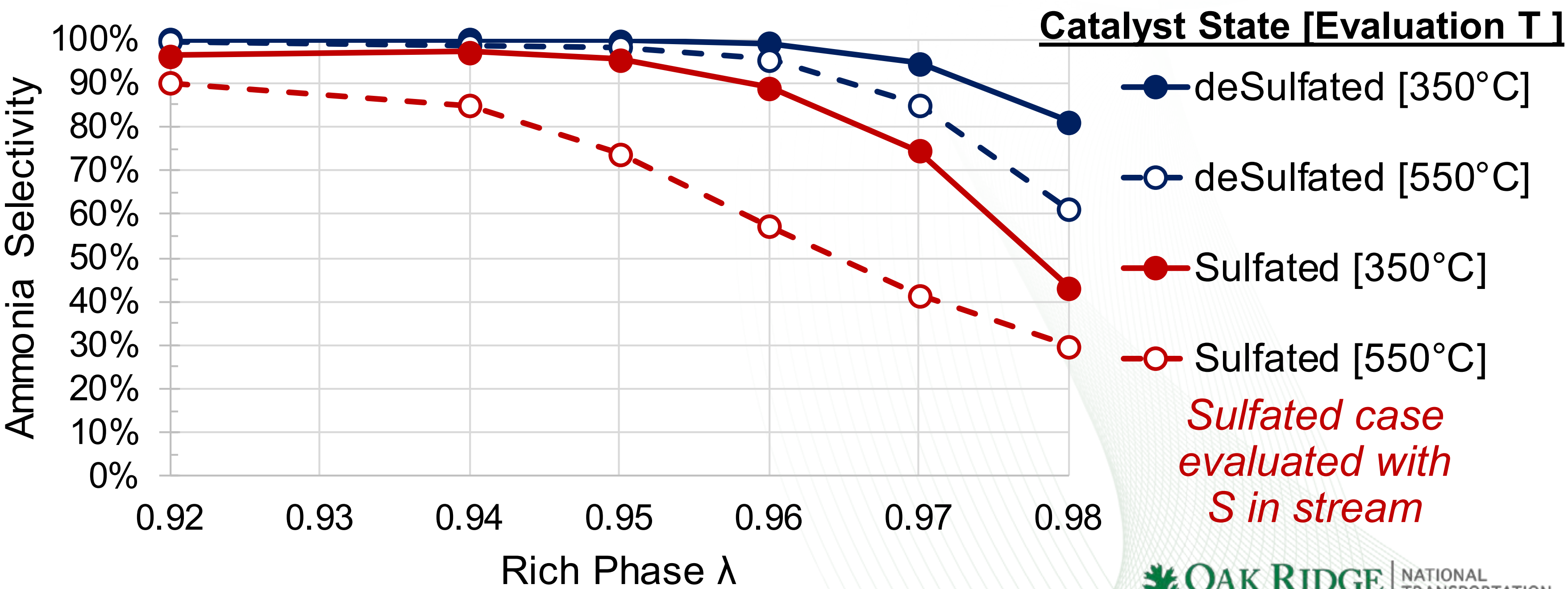
- Conducted passive SCR cycles on a synthetic exhaust gas flow reactor with 3 small pore Cu zeolite formulations
- Zeolite structure has minimal performance impact in middle of SCR T window (250-350 °C)
  - formulation effects observed at marginal Ts (≤200 °C, ≥400 °C)
  - formulations exhibit low vs. high T performance tradeoffs
- Rich timing (NH_3 dose, SCR NH_3 capacity utilization) has a strong effect
  - short rich times result in low NOx conversions (NH_3 coverage too low)
  - long rich times increase NH_3 slip and fuel penalty
  - optimal timing depends on T



S aging during lean-rich cycling at varying λ shows TWC is still capable of NH_3 production after aging

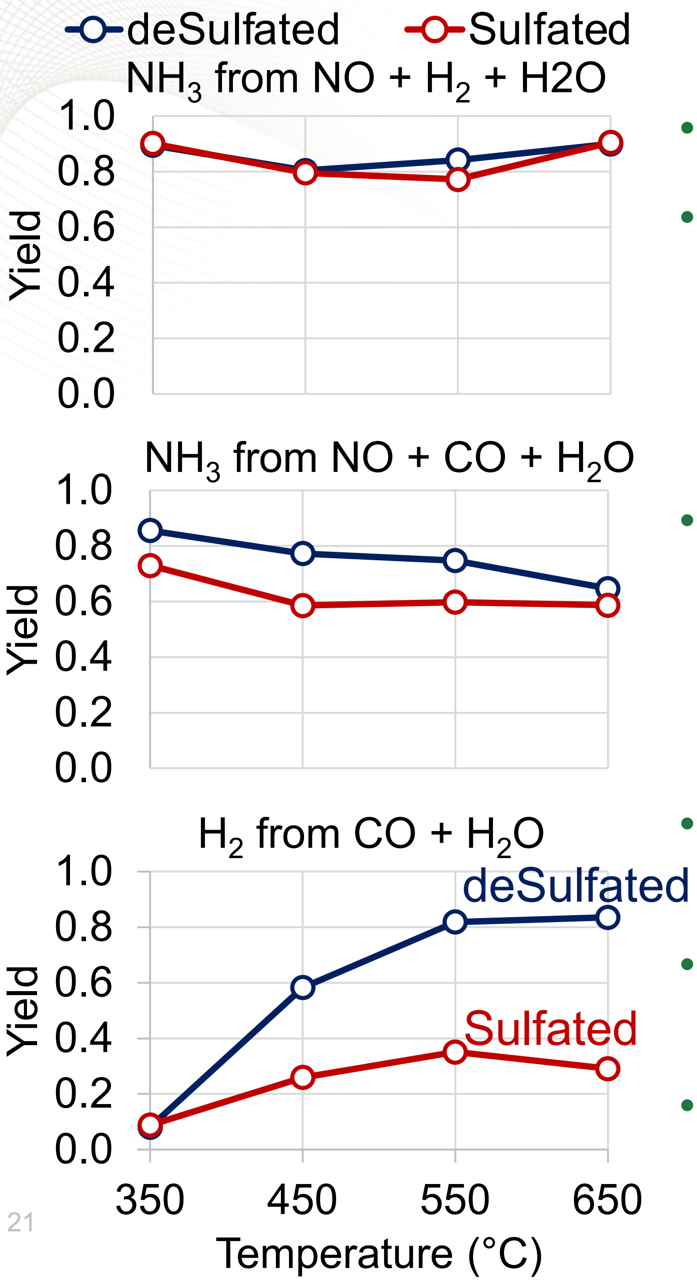
sample ID	Description	Pt (g/l)	Pd (g/l)	Rh (g/l)	OSC	NSC
Malibu-1	Front half of TWC	0	7.3	0	N	N

- Before evaluation:
  - Hydrothermally aged for 100 hours at 920°C
  - Exposed to 2ppm SO_2 for 12.5 hours under cycling conditions
- "Sulfated": after hydrothermal aging and S exposure under cycling conditions
- "deSulfated": after deSulfation at 650°C with lean-rich cycling for 3 hours



Sulfated case evaluated with S in stream

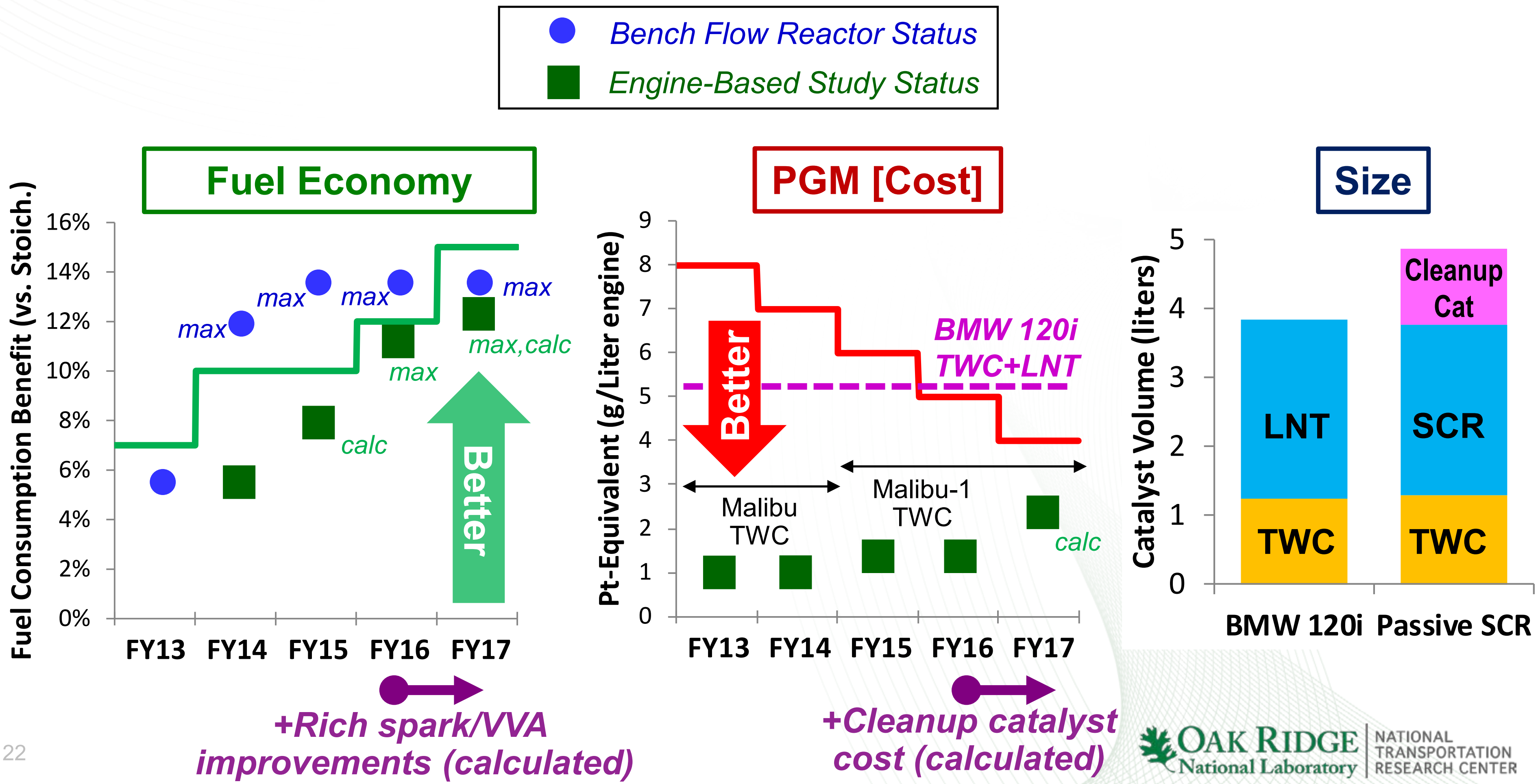
Analysis of S aged TWC shows NH_3 production even when water gas shift poisoned



- Isolating effects of sulfur on individual reactions
- Using H_2 as reductant leads to consistent activity before and after sulfation
  – Not primary means of catalyst deactivation
- Using CO as reductant leads to deactivation in NH_3 production
  – Still maintains high activity
- Complete water gas shift reaction to H_2 is heavily deactivated
- NH_3 production using CO even when WGS reaction is not active
- Molecular H_2 can be utilized but is not necessary for NH_3 production on catalyst

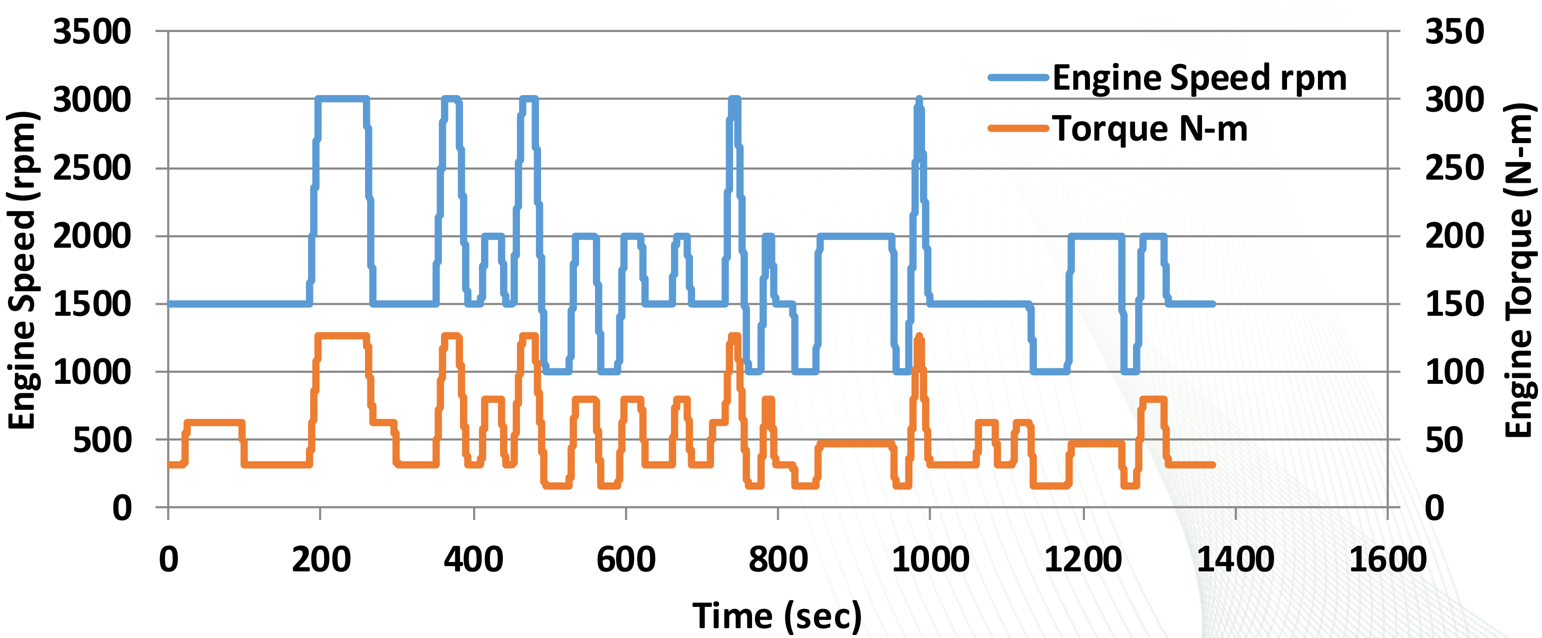
Remaining Challenges

- Transient drive cycle operation: combine knowledge gained to date to demonstrate performance over transient drive cycle or modal simulation
- HC/CO clean-up: demonstrate cleanup catalyst function/performance
- Further SCR studies: alternate formulations, aging



Future Work

- **FY17:** Remaining focus on annual milestone and engine system studies with 6-mode cycle recommended by GM for estimating transient drive cycle results
  - Measure emissions including N_2O
  - Measure fuel efficiency gain vs. stoichiometric GDI (calculate deS/desoot)
  - Include (2) load-speed points relevant to USDRIVE ACEC Tech Team Engine Efficiency Goals as well
- **FY18:** Reassess after FY17 milestone status to prioritize FY18 focus areas



Future work subject to change based on funding levels

Summary

- **Relevance**
  - Lean GDI engine emission control enables potential 10-15% fuel efficiency gain for gasoline-dominant U.S. light-duty fleet
- **Approach**
  - Bench flow reactor, engine, and aging studies are combined to study fuel efficiency and emissions relative to Tier 3 standard
- **Technical Accomplishments**
  - Bench Flow Reactor: Three Cu-SCR formulations feasible for passive SCR
  - Engine Studies: (1) Variable valve actuation and other controls give fuel efficiency gain, and (2) TWC with NOx storage component shows promise for short rich event NH_3 production
  - Aging: (1) Accelerated aging studies show effect of rich air-to-fuel ratio on ammonia production before and after sulfation under cycling conditions and (2) reaction probe experiments show effect of sulfur on different reaction pathways for NH_3 production
- **Collaborations**
  - GM, Umicore, and the University of South Carolina are primary partners
- **Future Work** *(subject to change based on funding levels)*
  - Focus on FY17 annual milestone based on GM-recommended 6-mode test
  - Continue addressing control challenges and pathways to maximize fuel efficiency

Technical Back-Up Slides

Project Goals Defined by Industry

In addition to milestones, a set of project goals has been adopted to ensure progression towards goal of low-cost emissions control solution for fuel efficient lean-burn gasoline vehicles

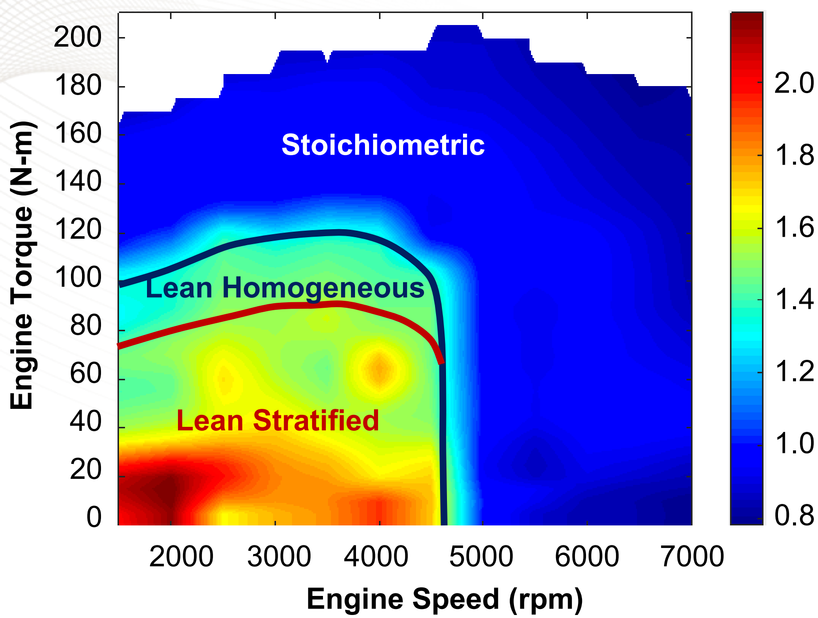
	FY13	*FY14*	*FY15*	*FY16*	*FY17*
Fuel economy gain over stoichiometric	7%	10%	10%	12%	15%
Total emissions control devices Pt* (g/L_{engine})	8	7	6	5	4

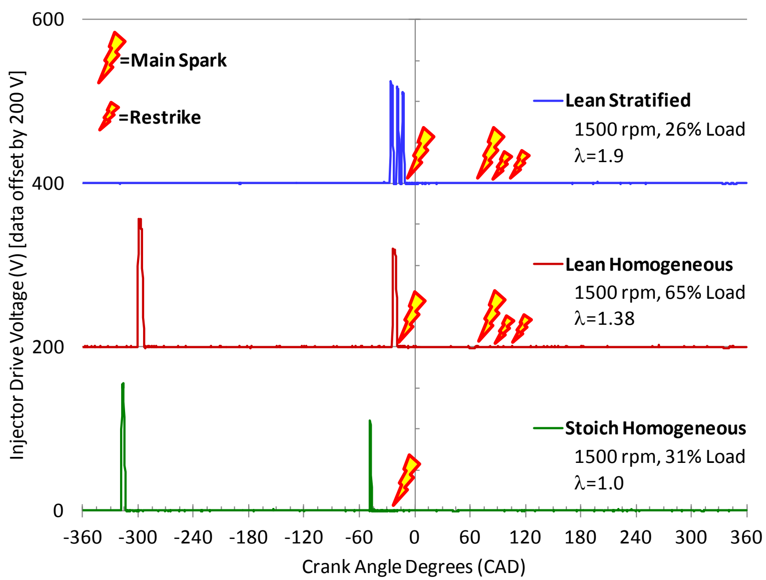
	5-year Average ($/troy oz.)	Pt-equivalent
Platinum	$ 1,504 /troy oz.	1.0
Palladium	$ 463 /troy oz.	0.3
Rhodium	$ 3,582 /troy oz.	2.4
Gold	$ 989 /troy oz.	0.7

* - will use Pt equivalent cost to account for different costs of Pt, Pd and Rh; 5-year average value fixed at beginning of project

As a reference point, the BMW 120i vehicle with a Euro 5 compliant TWC+LNT system contains a Pt-equivalent total of 5.1 g/liter of engine displacement

BMW 120i engine features three main combustion modes





- Center mounted combustion system design
- Lean Stratified
  - fuel injections close to TDC
  - multiple spark events
  - lambda ranges between 1.6 and 2.2
  - limited to 4500 rpm and 55% load

- Lean Homogeneous
  - two injections: one during intake stroke and one late in compression stroke close to TDC
  - multiple spark events
  - λ ranges between 1.4 and 1.6
  - limited to 4500 rpm and 55-75% load
- Stoichiometric
  - two injections: one during intake stroke and a smaller one early in compression stroke
  - single spark event
  - λ=1
  - entire engine operating range

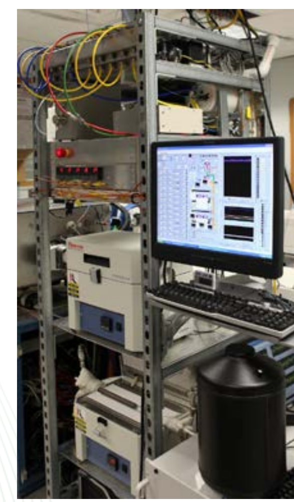
Conducted transient flow reactor experiments to estimate TWC effects on fuel consumption

- Used feedback-controlled cycles on flow reactor to evaluate dynamic TWC response in context of passive SCR
- Evaluated two different simulated engine cycles (fixed load, load step)

	fixed load		load step	
	rich	lean	rich	lean
load (BMEP)	2 bar	2 bar	8 bar	2 bar
SV (h^{-1})	**27000**	45000	**60000**	45000
NOx (ppm)	**600**	360	**1200**	360
max lean time	**50%**		**80%**	
simulates	cruise		“hill” transient	

	Rich						Lean
λ	0.95	0.96	0.97	0.98	0.99	1.00	2
O_2 (%)	0.96	1.02	1.07	1.13	1.17	1.22	10
CO (%)	2.0	1.8	1.6	1.4	1.2	1.0	0.2
H_2 (%)	1.0	0.9	0.8	0.7	0.6	0.5	0
NO (ppm)	600 (or 1200)						360
C_3H_8 (ppm C_1)	3000						1900
H_2O (%)	11						6.6
CO_2 (%)	11						6.6
TWC SV (hr^{-1})	27000 (or 60000)						45000

- Compositions & flows selected to mimic BMW GDI engine exhaust
- Space velocity changed with λ and load
- C_3H_8 chosen as challenging HC



Three-Way Catalyst (TWC) Sample Matrix

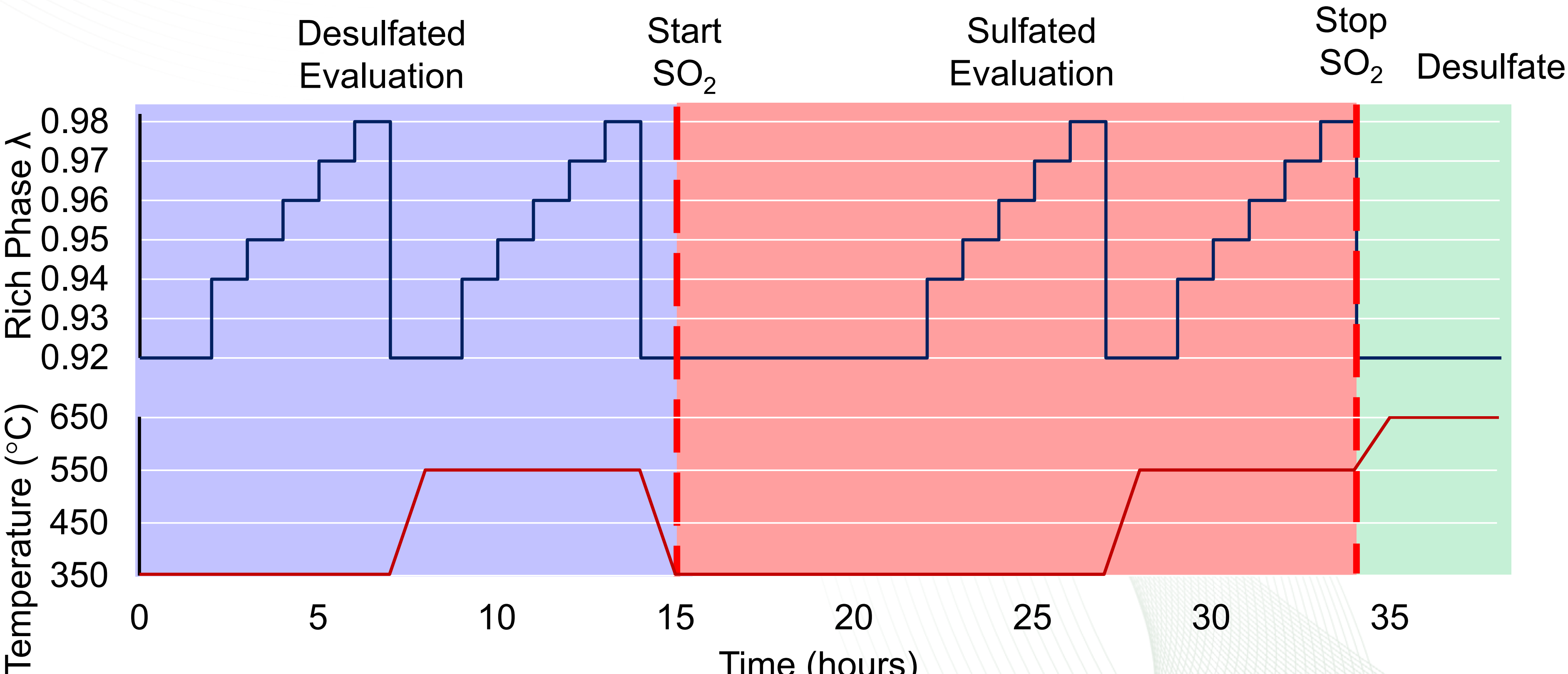
- **“Malibu” TWCs:**
  - Commercial state-of-the-art TWC from a MY2009 Chevrolet Malibu SULEV vehicle
- **“ORNL” TWCs:**
  - Prototype formulations supplied by Umicore specifically for this project

Catalyst Sample Matrix [OSC=oxygen storage capacity; NSC=NOx storage capacity]

sample ID	Description	Pt (g/l)	Pd (g/l)	Rh (g/l)	OSC	NSC
Malibu-1	Front half of TWC	0	7.3	0	N	N
Malibu-2	Rear half of TWC	0	1.1	0.3	Y	N
Malibu-combo	Full TWC	0	4.0	0.16	Y	N
ORNL-1	Pt + Pd + Rh	2.47	4.17	0.05	Y	Y
ORNL-2	Pd + Rh	0	6.36	0.14	N	N
ORNL-6	Pd	0	6.50	0	N	N
ORNL-5	Pd + OSC high	0	6.50	0	H	N
ORNL-4	Pd + OSC med	0	4.06	0	M	N
ORNL-3	Pd + OSC low	0	1.41	0	L	N

Rich λ Sweep Procedure for TWC S Aging

- Rich λ controlled through O_2, CO, and H_2 concentrations
  - Range: 0.92-0.98
- 350°C and 550°C tested
- Desulfated by cycling at 650°C for 3 hours.



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Reaction Probe Procedure: After S Aging and deSulfation

- H_2 production and NH_3 production measured at 350C, 450C, 550C, and 650C
  - Both desulfated and sulfated
- Equivalent reduction capacities used for different reductants
- Hydrogen production calculated as fraction of equivalent reductant
- Cycling 2 minutes lean, 2 minutes rich, rather than using LabVIEW feedback

	Cycled between 10% O_2 and N_2 balance SV = 27,000 hr^{-1}					
	CO + H2O		NO + H2 + H2O		NO + CO + H2O	
	Rich	Lean	Rich	Lean	Rich	Lean
CO (%)	1.0	1.0	0	0	1.0	1.0
H_2 (%)	0	0	1.0	1.0	0	0
NO (%)	0	0	0.05	0.05	0.05	0.05
C_3H_8 (%)	0	0	0	0	0	0
H_2O (%)	5.0	5.0	5.0	5.0	5.0	5.0
O_2 (%)	0	10.0	0	10.0	0	10.0